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BEFORE THE PATENT TRIAL AND APPEAL BOARD

Ex parte BETHANIE J.H. STADLER,
KOTHA SAI MADHUKAR REDDY,
DOUGLAS A. REKENTHALER, and PATRICK MCGARY[1]

Appeal 2015-007866
Application 13/309,673
Technology Center 1700

Before: CATHERINE Q. TIMM, KAREN M. HASTINGS, and AVELYN M. ROSS, *Administrative Patent Judges*.

TIMM, *Administrative Patent Judge*.

DECISION ON APPEAL[2]

STATEMENT OF CASE

Pursuant to 35 U.S.C. § 134(a), Appellants appeal from the Examiner's decision to reject claims 1, 26, and 27 under 35 U.S.C. § 103(a)

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[1] Appellants are the real parties in interest.

[2] In explaining our Decision, we cite to Final Office Action dated March 14, 2014 (Final), the Appeal Brief dated April 15, 2015 (Appeal Br.), the Examiner's Answer dated June 30, 2015 (Ans.), and the Reply Brief dated August 31, 2015 (Reply Br.).

as obvious over McGary[3] in view of Aksu[4] and Takeda.[5] We have jurisdiction under 35 U.S.C. § 6(b).

We AFFIRM.

The claims are directed to a method of electroplating an alloy of iron (Fe) and gallium (Ga) (Galfenol alloy) onto a substrate. *See, e.g.*, claim 1. The dispute is focused on the last clause of claim 1. *Compare* Final 5–8, *and* Ans. 6–13, *with* Appeal Br. 3–6, *and* Reply Br. 1–2. Claim 1, which we reproduce from the Claims Appendix of the Appeal Brief highlighting the portion of the claim at issue, reads:

> 1. A method of electro-plating a Galfenol alloy onto a substrate, comprising:
>
> providing an electroplating bath comprising sodium citrate and a mixture of Fe and Ga salts;
>
> providing a substrate in the electroplating bath; and
>
> providing a current in the electroplating bath to deposit Galfenol ($Fe_{1-x}Ga_x$, where x is in a range of from 10% to 40%) onto the substrate;
>
> *wherein the Fe^{2+}:Ga^{3+} ratio is between about 1:3-1:2, the amount of sodium citrate is equal to or less than that of Ga^{3+}, and the pH is between about 3-6, in the electroplating bath.*

Appeal Br. 8 (emphasis added).

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[3] McGary, "Electrochemically Synthesized Magnetic Nanowire Heterostructures and Arrays for Acoustic Sensing," Dissertation UMI Number 3316145 (2008).
[4] Aksu et al., US 7,507,321 B2, issued Mar. 24, 2009.
[5] Takeda et al., JP 2008-260981 A, published Oct. 30, 2008 (as translated).

As Appellants do not argue any claim apart from the others, we select claim 1 as representative to resolve the issue on appeal.

OPINION

McGary electroplates Galfenol alloy using an electroplating bath containing Fe^{2+}, Ga^{3+}, and sodium citrate, but as pointed out by Appellants, McGary does not disclose concentrations within the ratios of claim 1 along with a pH within the 3–6 range also required by claim 1. Appeal Br. 3. The issue is: Have Appellants identified a reversible error in the Examiner's determination that those of ordinary skill in the art would have had a reasonable expectation that baths with compositions meeting the concentrations and pH of claim 1 would have been suitable for electroplating a Galfenol alloy given the known effects of varying the concentrations and pH and the routine experimentation that would have been conducted to optimize the bath for the desired electroplating?

Appellants have not identified such an error.

McGary is a dissertation describing a project whose ultimate goal is to develop magnetostrictive nanowires that can be used as acoustic sensors. McGary § 3.1 at pp. 40–41. In essence, the project gets its inspiration from the cilia in the ear and seeks to create a biomimetic smart structure that oscillates with incoming acoustic waves in a way that mimics those cilia. McGary § 1.1.1 at p. 2; § 1.3 at p. 6.

McGary started the project by developing an electroplating bath for Galfenol. McGary Chap. 7 at p. 175. Chapter 3 describes the experiments conducted to develop the bath chemistry. The product of these experiments was a bath able to electroplate metallic films of $Fe_{1-x}Ga_x$ ($12<x<27$ at%)

using several formulations of Fe and Ga sulfate with sodium citrate as a complexing agent. McGary Chap. 7 at p. 175. There is no dispute that McGary discloses a method of electroplating a Galfenol alloy onto a substrate according to the providing steps of claim 1 using solutions including Fe^{2+}, Ga^{3+}, and sodium citrate in various concentrations, some of which meet the concentration limitations of claim 1. *Compare* Final 5–6, *with* Appeal Br. 6–13.

McGary first conducts experiments to determine a suitable complexing agent for a bath using $FeSO_4 \cdot 7H_2O$ (Fe (II) sulfate) and $Ga_2(SO_4)_3 \cdot 18H2O$ (gallium (III) sulfate) as sources of Ga^{3+} and Fe^{2+} ions, respectively. McGary § 3.3.1 at p. 45. McGary selects sodium citrate as a suitable complexing agent for Ga^{3+}, and then conducts experiments to develop an optimized process for controlling the stoichiometry of the electroplated alloys. McGary § 3.2.1 at p. 42:7–11. The experiments involved testing different citrate-containing baths to find optimum electroplating conditions for obtaining $Fe_{1-x}Ga_x$ (12<x<30 at%). McGary § 3.3.1 at p. 45.

Table 3.2 details four series of tested baths. *Id.* To the series PI bath, ascorbic acid and/or sodium D-gluconate were added. McGary § 3.3.1 at pp. 45–46; *see also* Table 3.2. In the series A baths, the amount of Fe^{2+} was varied, which varied the Fe^{2+};Ga^{3+} ratio. McGary § 3.3.1 at p. 46; *see also* Table 3.2. In the series B baths, the Ga^{3+} and citrate concentrations were again held constant, but at higher concentrations, and the Fe^{2+} concentration again varied. *Id.* In the series C baths, which are the focus of the Examiner's rejection and Appellants' arguments, the Fe^{2+} concentration was held constant at 0.04 M, the Ga^{3+} concentration was held constant at 0.1 M,[6] and sodium citrate was varied in 0.05 M increments between 0.05-0.25 M. *Id.*

The reason McGary performed the series C tests was because it was determined that the citrate concentration seemed to impact the metal distribution in the electroplated $Fe_{1-x}Ga_x$ film and the current efficiency of the electroplating process. McGary § 3.5.2 at p. 65. McGary shows the resulting metal strips that were electroplated using a Hull Cell with the series C baths in Figure 3.2. McGary measured the pH with each 0.05 M addition of citrate and reports the result in Figure 3.2. Taking the information from Table 3.2 and Figure 3.20, the values of interest are as follows:

ID (Table 3.2/Fig. 3.20)	Fe^{2+}:Ga^{3+}	Fe^{2+} conc.	Ga^{3+} conc.	sodium citrate conc.	pH
C1/(a)	1:2.5	0.04 M	0.1 M	0.05 M	2.0
C2/(b)	1:2.5	0.04 M	0.1 M	0.10 M	2.1
C3/(c)	1:2.5	0.04 M	0.1 M	0.15 M	2.3
C4/(d)	1:2.5	0.04 M	0.1 M	0.20 M	3.0
C5/(e)	1:2.5	0.04 M	0.1 M	0.25 M	4.5

According to McGary, C1 (pH=2) only resulted in a very small region of a brittle metallic film. McGary § 3.5.2 at p. 65. McGary indicates that better results were obtained with C2, C3, and C4 solutions. McGary § 3.5.2 at pp. 65–66. According to McGary,

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[6] Because each gallium (III) sulfate molecule generates two Ga^{3+} ions, the concentration of Ga^{3+} ions is twice the gallium (III) sulfate concentration. McGary § 3.3.1 at p. 45. Thus, the 0.05 M gallium (III) sulfate concentration listed for the series C concentration in Table 3.2 is equal to 0.1 M Ga^{3+}.

> a bright region was visible in C2 (pH=2.1), and the metal distribution continued to increase with added citrate concentration. The composition results are displayed in Figure 3.21 for solutions C2, C3, and C4. In each case, the films containing more than 25% Ga were oxidized significantly. With increasing pH, the bright range, however, was shifted to the lower current densities. Solution C3 showed the most promising Ga range. The overall current efficiency was also measured gravimetrically for each test and increased from 6% in C2 to 10% in C4, but reduced slightly in C5 to 9.5%.

McGary § 3.5.2 at pp. 65–66.

The Examiner focuses on the C2/(b) bath composition. Final 6. This bath includes a Fe^{2+}:Ga^{3+} ratio (1:2.5) within the range of claim 1 (1:3–1:2) and an amount of sodium citrate (equal to that of Ga^{3+}) within the range of the claim (equal to or less than that of Ga^{3+}), but the C2(b) bath does not have the required pH of between about 3–6. *See* table above.

Although McGary reports the pH for the C series baths, McGary does not disclose that the bath is optimized for pH outside of adjusting the citrate concentration. However, McGary § 3.2.1 makes clear that pH was a concern in the electroplating process and that a pH buffer was needed ameliorate the effects of local depletion of H^+ ions on the cathode that likely causes non-adherent gallium oxide on the cathode. McGary § 3.2.1 at pp. 41–42. This section also explains that early work on the electrolytic refinement of Ga was performed in both a basic sulfate solution of potassium hydroxide (KOH) and a solution of gallium hydroxides in dilute sulfuric acid. McGary § 3.2.1 at p. 42. McGary notes that the early work, however, resulted in globules or trees. *Id.*

Takeda provides further background, which informs us of the knowledge within the art with regard to pH and its effects on gallium plating. Takeda discloses a process in which

> the gallium plating solution contains a chelating agent, and the pH is set in the range of 3 to 10. In a conventional plating solution, gallium hydroxide is formed close to the neutral zone that generates turbidity and precipitate in the plating solution. For this reason, the current efficiency deteriorated remarkably or this hydroxide was incorporated into the plating film and the membranous quality became drastically poor, which caused an obstacle to application on an industrial level.
>
> In the present invention, by adding a chelating agent, even in a pH range of 3 to 10, decrease in current efficiency and deterioration of membranous quality can be prevented.

Takeda ¶¶ 29, 30.

Aksu also discusses the interplay between pH, H^+ species, and Ga oxides and hydroxides in gallium electroplating and the prior art use of NaOH and KOH to raise pH. Aksu col. 2, ll. 17–44. Aksu explains that electroplating Ga out of low pH baths may suffer from low cathodic efficiencies arising from the presence of a large concentration of H^+ species. Aksu col. 2, ll. 17–44. But, as further explained by Aksu, as the pH is increased, Ga forms oxides and hydroxides, which may precipitate. *Id.* It is possible to electroplate in a bath of very high pH (pH>14) using high concentration of KOH and NaOH, but such high alkalinity is corrosive to equipment and has high viscosity. *Id.* Aksu's inventive bath uses citrate as a complexing agent and adjusts pH using, for instance, NaOH to obtain bath pHs within the range of 7 or higher, the pH ranges being sometimes significantly higher such as between 10 and 13. Aksu col. 4, ll. 33–53; col. 5, ll. 8–12 (Example 1).

All of the references evince a knowledge in the art that the pH was a result effective variable in the process of electroplating gallium and McGary provides evidence that this holds true for electroplating of $Fe_{1-x}Ga_x$ (12<x<30 at%) as well. The evidence as a whole also provides evidence that adjusting the pH to levels within the 6–9 range of the claim was known to provide adequate electroplated films. The use of KOH and NaOH as well as combinations of those caustics with citrate as a complexing agent to adjust pH was known in the art. On this record, there is sufficient evidence to shift the burden to Appellants to show that the bath composition in the pH of claim 1 results in $Fe_{1-x}Ga_x$ electroplated films with unexpected properties. *See In re Aller*, 220 F.2d 454, 456 (CCPA 1955) (explaining that where the general conditions of a claim are disclosed in the prior art, and the evidence supports a determination that discovery of the optimum or workable ranges would result from routine experimentation, the burden shifts to the applicant to show that the particular range values recited in the claim produce an unexpected result). Appellants do not rely upon a showing of unexpected results on this record.

CONCLUSION

We sustain the Examiner's rejection.

DECISION

The Examiner's decision is affirmed.

TIME PERIOD FOR RESPONSE

No time period for taking any subsequent action in connection with this appeal may be extended under 37 C.F.R. § 1.136(a)(1).

AFFIRMED